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LASER PHYSICS AND LASER TECHNIQUES.(U)

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01 January 1976 - 31 December 1976

LASER PHYSICS AND LASER TECHNIQUES

Principal Investigator

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Department of Electrical Engineering

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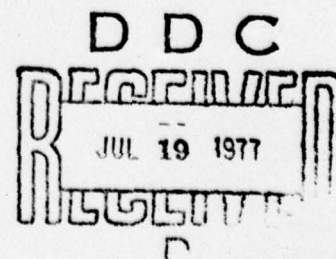
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ABSTRACT

Substantial improvements have been made in the performance of the optically pumped 546.1 nm Hg laser. DC-pumped operation of sealed-off laser tubes has been demonstrated with no apparent lifetime limitations. Several new pump lamp designs have been developed and tested to overcome limitations set by emission line self-reversal, cataphoresis, and single-isotope mercury fill requirements. A novel dc-excited "diffusion-bypass" lamp has been shown to provide a particularly effective pumping source. Extensive measurements of gain, saturation intensity, and power output have been made, with both natural and single-isotope mercury. A detailed theoretical analysis of pumping and relaxation processes in the Hg laser has been developed for interpreting these experimental measurements. Laser operation with Ar-H₂⁰-Hg and He-H₂⁰-Hg gas mixes in addition to the previous N₂-Hg gas mix has been obtained but found to be less efficient. Preliminary observations of line overlap between the mercury laser and various molecular absorbers for absolute wavelength stabilization have been made by observing mercury-laser-induced fluorescence in Br₂, I₂¹²⁷ and I₂¹²⁹ cells, and by making preliminary observations of a saturated-absorption feature in I₂¹²⁷ using a single-frequency Hg²⁰² laser. The optically pumped mercury laser continues to appear interesting as an absolute wavelength stabilized visible laser, as a potential ring laser gyroscope medium, and as a special-purpose low-power visible green laser.



RESEARCH ACCOMPLISHMENTS

The research carried out under this contract during the period 01 January 1976 to 31 December 1976 has been focused on improving the experimental performance and the theoretical understanding of the optically pumped 546.1 nm mercury laser first operated by Djeu and Burnham of the Naval Research Laboratory. This particular laser system appears to be promising for a number of practical applications, notably as a possible visible wavelength standard, as a laser medium for ring laser gyroscopes, and possibly as a special purpose low-power visible laser in the green. Significant technical accomplishments toward the understanding and improvement of this laser have been achieved during the current program, as outlined in more detail below.

At the time the original proposal for this research was submitted, a supplemental proposal was also submitted for a parallel program of more extensive research on aspects of the mercury laser system specifically aimed at the ring laser gyroscope application. This supplemental program was also funded and work on it initiated approximately halfway through the period of this contract. Work on this supplemental program is still continuing. Because of the close relationship of the technical work under the two programs, and because of the additional results and measurements that are expected to be completed under the supplemental program, it has seemed most useful to defer detailed publication of the full results from both programs until the supplemental program has also been completed. We expect therefore, that more

detailed results from this work and from the supplemental program will be published together in one or more scientific journal articles plus two Ph.D. dissertations to be prepared in mid-1977.

The research accomplishments under the present program have included the following:

1. DC-Excited Operation of the Mercury Laser

Figure 1 shows the two-step optical pumping process at 253.7 nm and 404.7 nm and the laser transition at 546.1 nm involved in the optically pumped Hg laser. In the original Djeu and Burnham version of this laser, a special annular rf-excited lamp requiring ~ 1 kW or more of rf input was employed to obtain ~ 2 mW output from a laser tube containing a flowing mixture of N₂ and Hg. Early in our program cw operation of this laser was obtained using two standard dc-excited low-pressure and low-power Hg lamps (inexpensive commercially available Hg germicidal lamps) in either a double-elliptical pump cavity or a close-wrapped pump configuration. Laser output of ~ 1 mW was obtained from a flowing laser tube with ~ 80 W total dc input to two such pump lamps, using the general arrangement illustrated in Fig. 2. This initial system was far from optimized but indicated the potential performance capabilities of this laser.

2. Sealed-Off Laser Operation

In the original Djeu and Burnham experiments and in many of our parametric studies the Hg laser tube was operated with a slowly but continuously flowing gas mixture of N₂ and Hg, since this permits experimental variation and optimization of both the N₂ and Hg pressures. However, we also processed and operated early in this program two fully sealed-off laser tubes, using careful

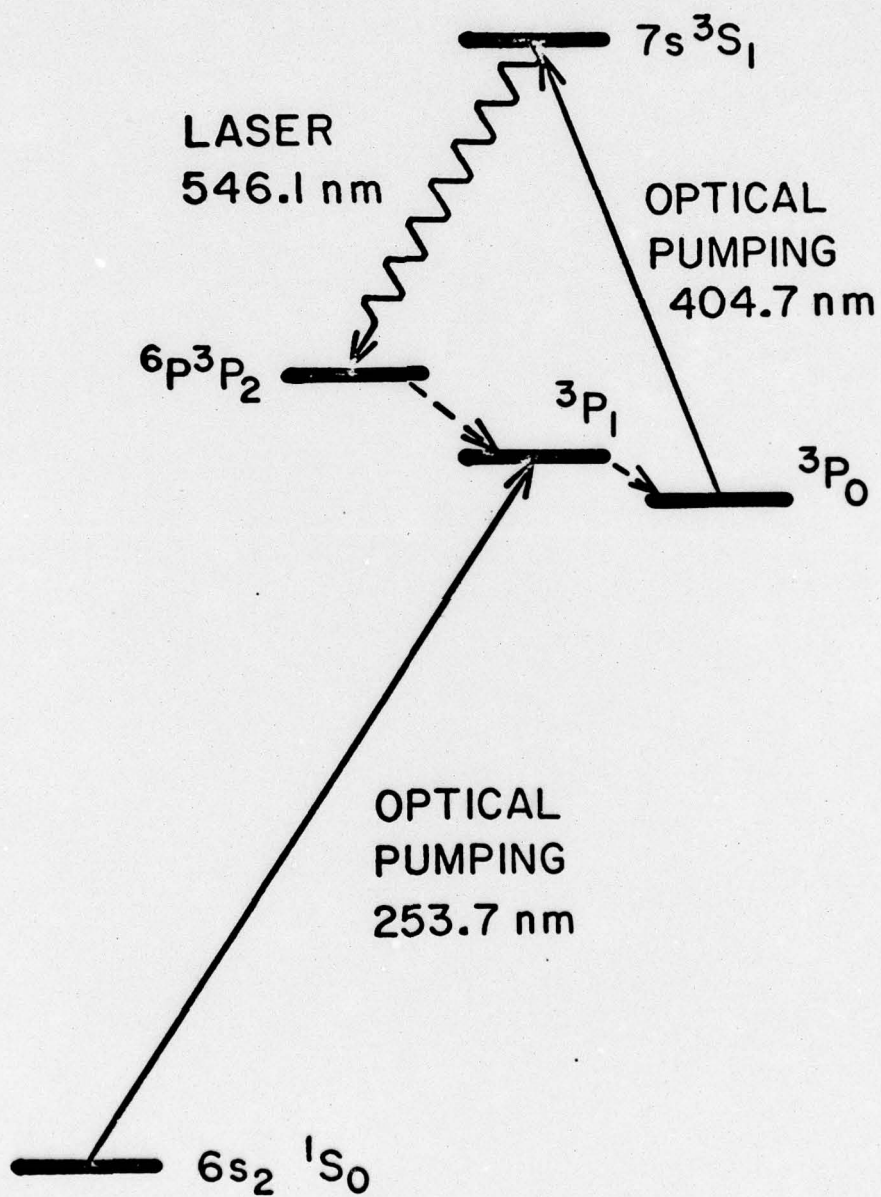
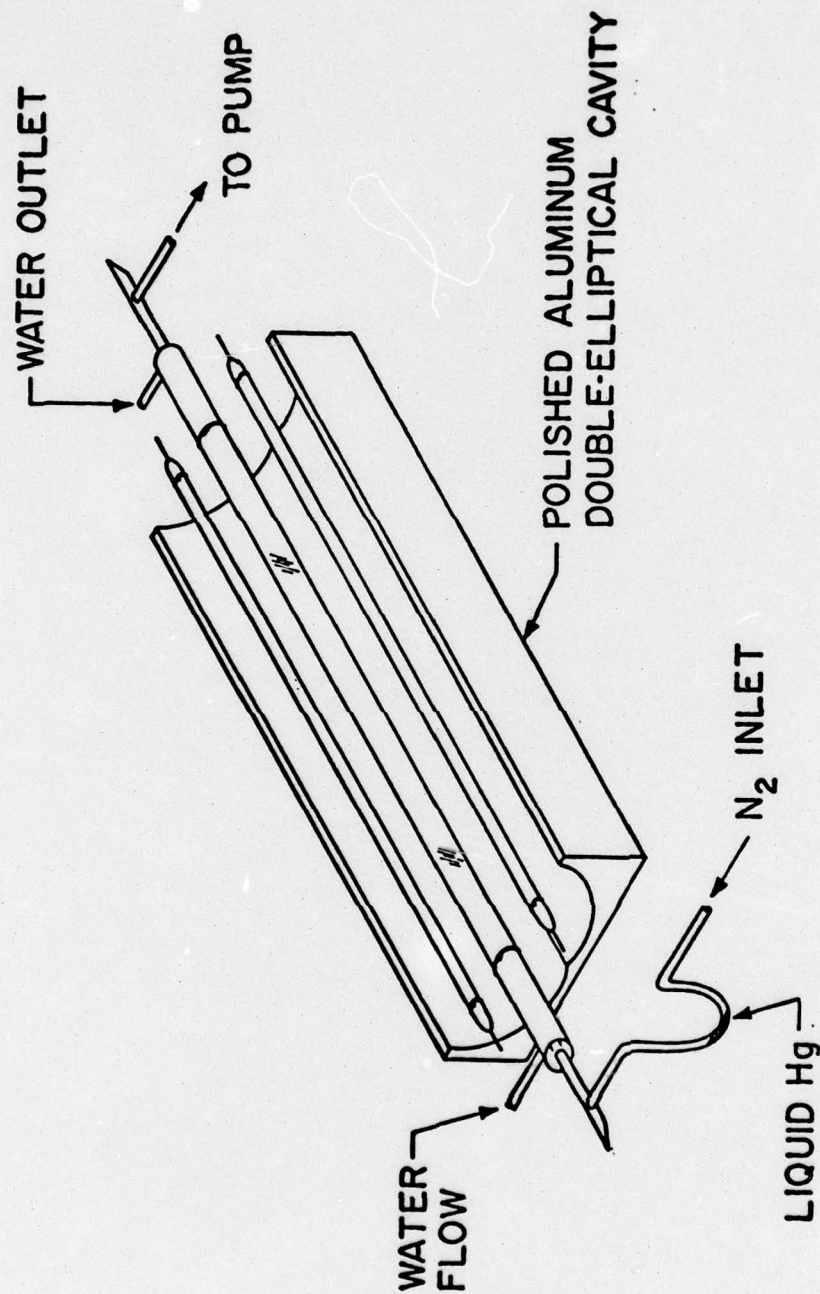


FIGURE 1



SCHEMATIC OF Hg-N₂ LASER EXPERIMENT

FIGURE 2

cleaning and processing procedures for the fabrication of the tubes. Laser outputs of ~ 2 mW with ~ 300 W input to a single low-pressure mercury arc lamp could be obtained from these tubes, even without optimizing all parameters. While extensive life tests were not carried out, no significant deterioration in the performance of these tubes was observed over a period of at least several months.

It has been suggested that the 3P_0 Hg metastables which are normally produced by the 253.7 nm pumping radiation in the Hg laser might alternatively be produced by a weak plasma discharge in the laser tube itself. We observed experimentally however that exciting an rf plasma discharge in one of the sealed-off laser tubes caused the rapid formation of a brown deposit on the tube walls. Formation of this compound, tentatively identified as an azide compound $Hg_3 N_2$, appeared to consume most of the gas in the tube. However, it was also found that gentle heating of the laser tube decomposed this compound, and restored normal operation of the laser.

We conclude in general that there should be no barrier to long-lived stable operation of sealed-off versions of this laser, provided that very careful clean processing techniques are employed at every stage in the tube construction.

3. Toronto Arc Pump Lamps

Mercury discharge lamps, either low-pressure glow discharges or higher-pressure arc discharges, can provide highly efficient sources of both ultraviolet and visible radiation. Mercury discharge have been the subject of extensive research for more than seventy years, and commercial mercury lamps are widely employed.

The Hg laser places very stringent requirements on its pumping source, however. The pump lamp must be simultaneously bright at both 253.7 and 404.7 nm, as well as essentially unbroadened if the pumping radiation is to be absorbed within the narrow absorption lines of the low-pressure Hg vapor in the laser tube. Efficient conversion from electrical input to radiative output from the pump lamp is obviously desirable for efficient laser pumping. At the same time the line broadening and self-reversal typically observed in mercury discharge lamps at higher currents and pressures must be avoided if the emitted radiation is to be useful for laser pumping. As a practical matter the lamps should be long-lived and require a minimal amount of mercury fill, especially if single-isotope operation is to be employed. The development of suitable pump lamps to meet these requirements formed a major portion of the research effort under this program.

In general terms, low-pressure and low-power mercury glow discharges can provide extremely efficient sources of unbroadened 253.7 nm radiation (>60% electrical efficiency in some cases). Such discharges are widely exploited in inexpensive commercial germicidal lamps. However, such lamps are much less bright and efficient at 404.7 nm. Also, increasing either the discharge current of the Hg vapor pressure tends to reduce efficiency, produce discharge instabilities, and/or to cause increasing self-reversal of the emitted radiation. Low to medium-pressure Hg arc lamps, by contrast can be much more efficient as sources of visible radiation, including 404.7 nm, but with greatly decreased 253.7 nm output and with serious self-reversal problems at both wavelengths.

An initial step in our lamp development efforts was the construction of a temperature-controlled, water-cooled-bore, liquid-pool-cathode, low-pressure arc as a pump lamp. Lamps of this type, often referred to as "Toronto arcs", were

widely used in earlier decades as bright and unbroadened line sources for Raman spectroscopy. With a single lamp of this type, having separate water-cooling circuits for the tube bore and for the mercury pool electrodes, ~ 3.5 mW of laser output was obtained with ~ 600 W input (1 A at 600 V) to the lamp. This performance could undoubtedly have been further improved. However, the complexity of this lamp plus the use of liquid-pool cathodes and the large volume of mercury required made other solutions seem more attractive, as described in the following.

4. Low-Pressure Hot-Cathode Mercury Arc Lamps

As a next step several low-pressure cooled-bore discharge lamps were operated using heated tungsten dispenser cathodes (Spectramat cathodes). These lamps typically contained ~ 300 mTorr of argon buffer gas plus an initial 20 mg fill of Hg, with the Hg vapor pressure controlled by varying the water-cooled bore temperature.

One of the first and most important results obtained with these lamps was that neither the tungsten dispenser cathode itself nor the free barium evaporated by this cathode seriously gettered or absorbed the small amount of mercury fill in the lamp. The lamp performance as a laser pump source was also very good. Sustained operation with ~ 7 mW output from a flowing laser setup was typically obtained with ~ 400 W input to lamps of this type.

The operating life of these lamps in a single session was, however, seriously limited by the large and unavoidable transfer of mercury along the lamp bore from anode to cathode by cataphoresis in the discharge. After typically 5 to 7 hours of operation the mercury fill was entirely transferred

and condensed into the cathode end of the lamp. Lamp operation could then be fully restored only by an ~ 4 hour low-temperature baking cycle to redistribute the mercury into the lamp bore.

5. Diffusion-Bypass Lamps

A long-term solution to the cataphoretic pumping problem was obtained by constructing a further series of lamps with essentially the same cathode structure and gas fill, but with a large-bore diffusion-bypass tube from the cathode to the anode end, forming a rectangular closed-loop lamp. Figure 3 illustrates the general structure of these lamps. The mercury vapor pumped into the cathode region by cataphoresis in these lamps is continuously recycled back to the anode end by diffusion through the large-bore diffusion-bypass tube. Metal honeycomb baffles in this tube block any discharge through the tube while permitting free diffusion of the Hg vapor.

This lamp appears to provide a satisfactory solution to all of the major pump lamp problems. Both natural and single-isotope lamps of this type have been built and operated. Cumulative operation over some 300 to 400 hours has been obtained from at least one such lamp without noticeable deterioration. We believe that this lamp design provides a very satisfactory general solution to the problem of pumping the mercury laser with long life and good efficiency.

6. Inductively-Coupled Radio-Frequency Lamps

As an alternative solution to the lamp problem we have also considered, but have not experimentally tested, the use of an inductively coupled closed-loop rf-discharge mercury lamp. A lamp of this type would be constructed in the form of an electrodeless rectangular closed loop, inductively excited as

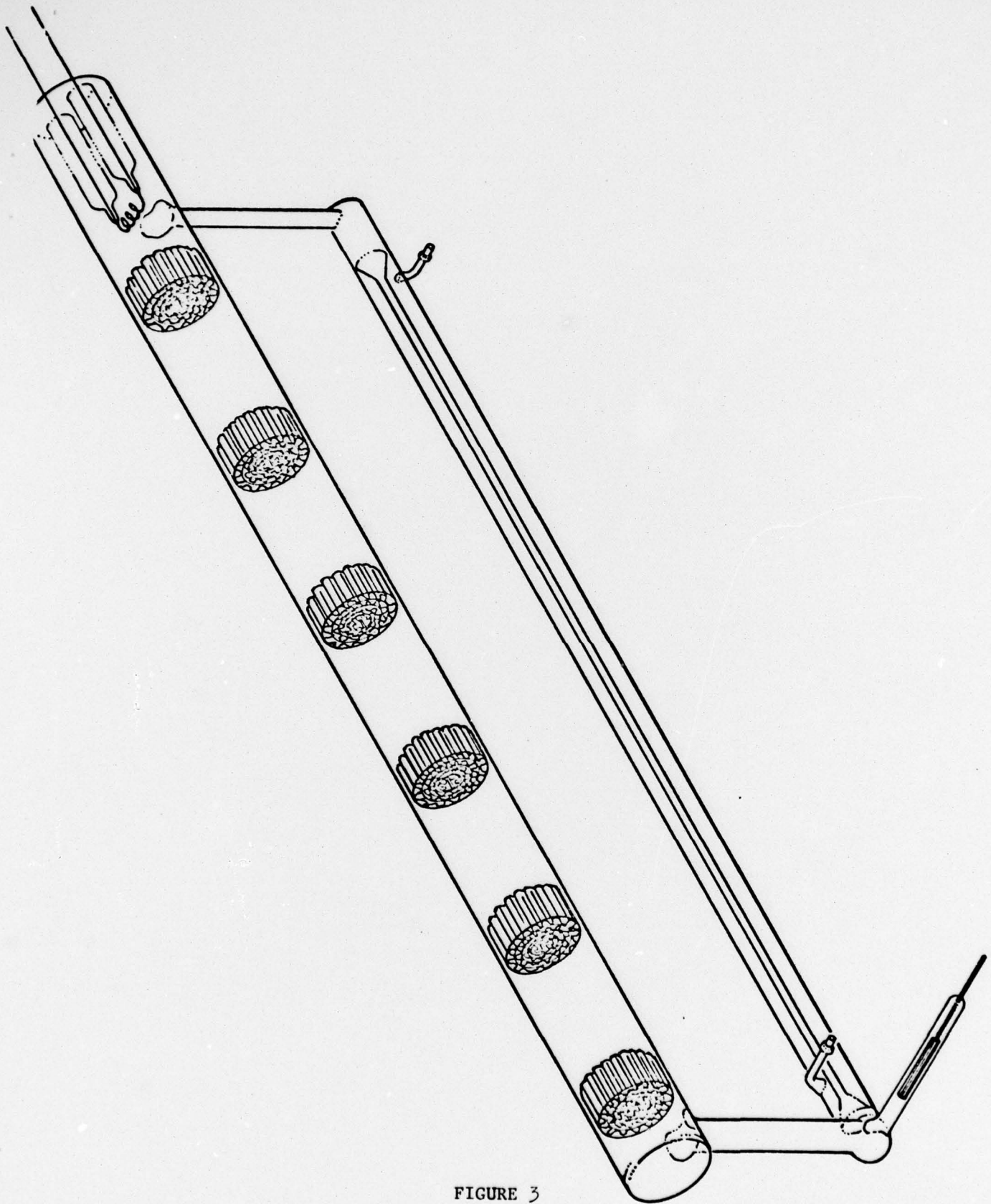


FIGURE 3

a single-turn secondary coupled to a primary excitation coil driven by a high-power rf oscillator in the 10 to 100 MHz range. The structure and method of excitation would thus be generally similar to the inductively coupled ion laser structures developed by Bell, Hodges and Goldsborough, and also to the inductively coupled xenon lamps used for Nd:YAG laser pumping by Huchital and Steinberg. Such a mercury lamp might use both long arms of a rectangular closed loop as radiation sources in double-elliptical or closed-wrapped pumping cavities, or only one arm might be used as a light source with the other arm providing only a large-bore low-impedance path to complete the closed loop.

We believe that a lamp of this type could provide some very significant advantages, starting with simple all-quartz construction requiring no electrodes or electrode seals. Cataphoresis, electrode problems, and gas cleanup should be essentially eliminated, and minimum mercury fill for single-isotope operation should be possible. The current distribution and the atomic excitation should be substantially more uniform across the bore diameter in an rf-excited lamp compared to a dc-excited lamp, producing increased brightness and decreased problems with self-reversal at higher mercury vapor pressures. Radio-frequency power supplies and rf discharge excitation in general can be efficient and stable, avoiding in particular the discharge instabilities characteristic of dc discharges. The only apparent practical difficulties with this approach center around the problems of impedance matching into the lamp at different power levels, and the necessity for suitable rf shielding around the overall laser structure. In general we believe this approach merits further consideration and possibly experimental testing in any further Hg laser development programs.

7. Single-Isotope Lamp and Laser Operation

Natural mercury consists of approximately 6 isotopes with significant abundance; and there are significant isotopic and hyperfine splittings of the levels involved in the laser pumping and oscillation processes. Figure 4 illustrates by way of illustration the hyperfine splitting of the 253.7 nm line of Hg. If the mercury isotopes are all essentially uncoupled in both the pump lamp and the laser-tube, one would expect a substantial performance improvement, by perhaps a factor or two or more, on going from natural mercury to a matched single isotopic mercury fill in both lamp and laser tube. Therefore, after carrying out the laser and lamp development experiments described thus far using natural mercury, we shifted to single-isotope lamp and laser experiments using the lowest-cost even isotope Hg^{202} . An even isotope is required to avoid nuclear hyperfine splitting effects, as illustrated in Fig. 4. Detailed experiments with the single isotope system are still continuing, and in general there is some continuing difficulty in obtaining precise and reproducible quantitative measurements, because of the sensitivity of both lamps and laser tubes to small impurities and processing variations. However, our tentative observation is that the performance improvement in going from natural to single-isotope Hg is only ~ 20% to 50% in gain and power output. Optimized single-isotope performance in a flowing-gas laser has been as high as ~ 13 mW output with ~ 350 W input to a single diffusion-bypass lamp. Further theoretical and experimental work is needed to understand the dynamics of isotopic energy transfer in the multi-isotope laser as compared to the single isotope case, and we hope to accomplish much of this in the remaining portion of the supplemental program.

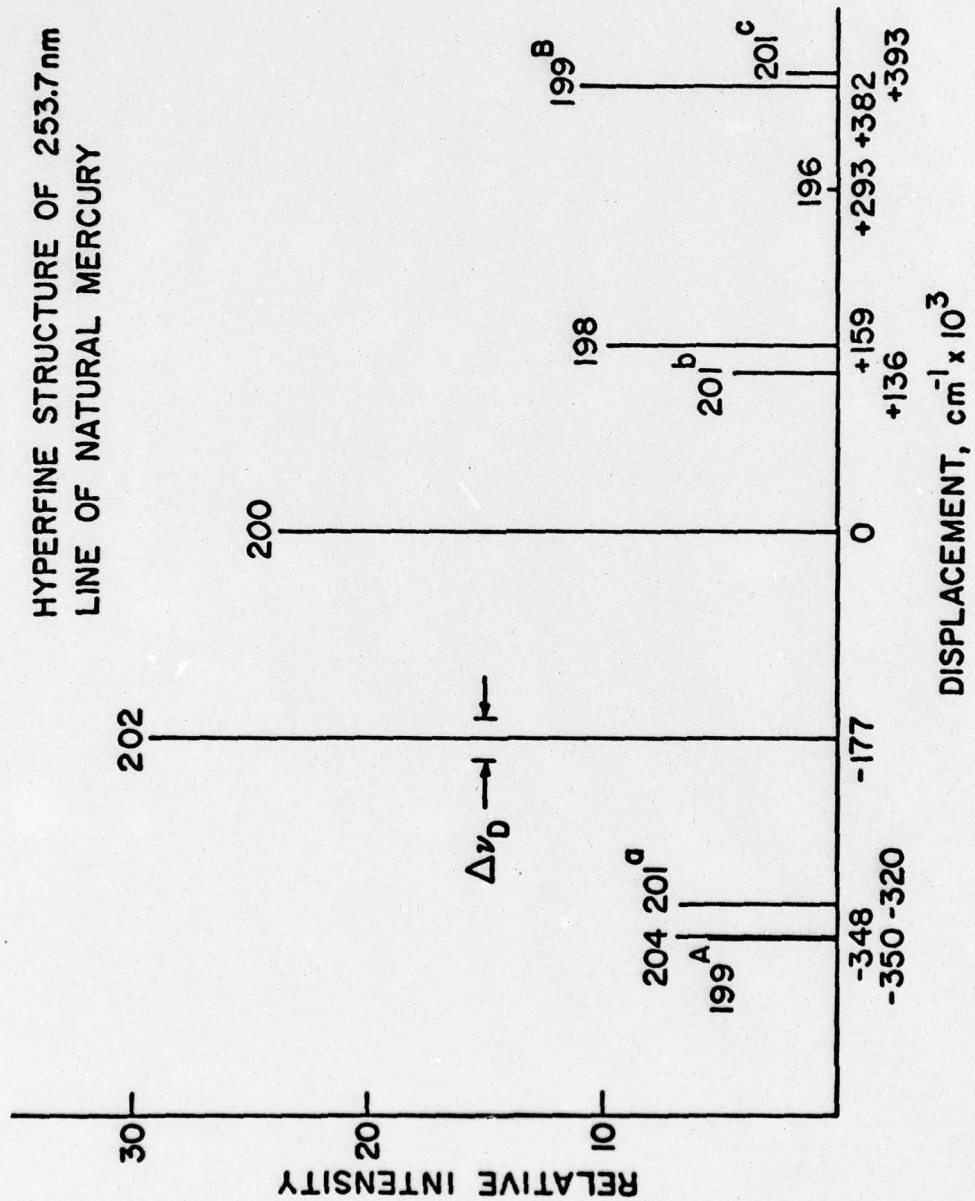


FIGURE 4

8. Gain and Saturation Intensity Measurements

In order to gain a quantitative understanding of the Hg laser performance we have carried out and are continuing to make extensive measurements of the parametric dependence of the small-signal gain, power output and saturation intensity of the Hg laser. Because of the difficulties in obtaining totally reproducible performance mentioned above, plus the additional measurements that are still in progress, we wish to defer detailed reporting and interpretation of these results until completion of the supplemental program. However our measurements have shown small-signal gains as high as $\sim 7\%$ per meter in a flowing single-isotope laser with a single pump lamp, and saturation intensities that are pressure-dependent but typically $\sim 1 \text{ W/cm}^2$. We expect to have much more detailed measurements and understanding of these parameters by the termination of the supplemental program. Figures 5 through 10 illustrate some of the preliminary measurements that have been made to date.

9. Theoretical Pumping and Rate Equation Analyses

Another major aspect of this program has been a detailed review of the previous literature on excitation and relaxation processes in nitrogen-mercury mixtures, in combination with a detailed rate equation analysis of the pumping, gain and relaxation processes in the mercury laser. Again, although very significant results have already been obtained from this effort, we prefer to delay a detailed report of these results until we can present a completed description at the finish of our continuing work.

In general, the existing literature on the relevant excitation and relaxation processes in excited Hg atoms is very extensive, but it is also neither

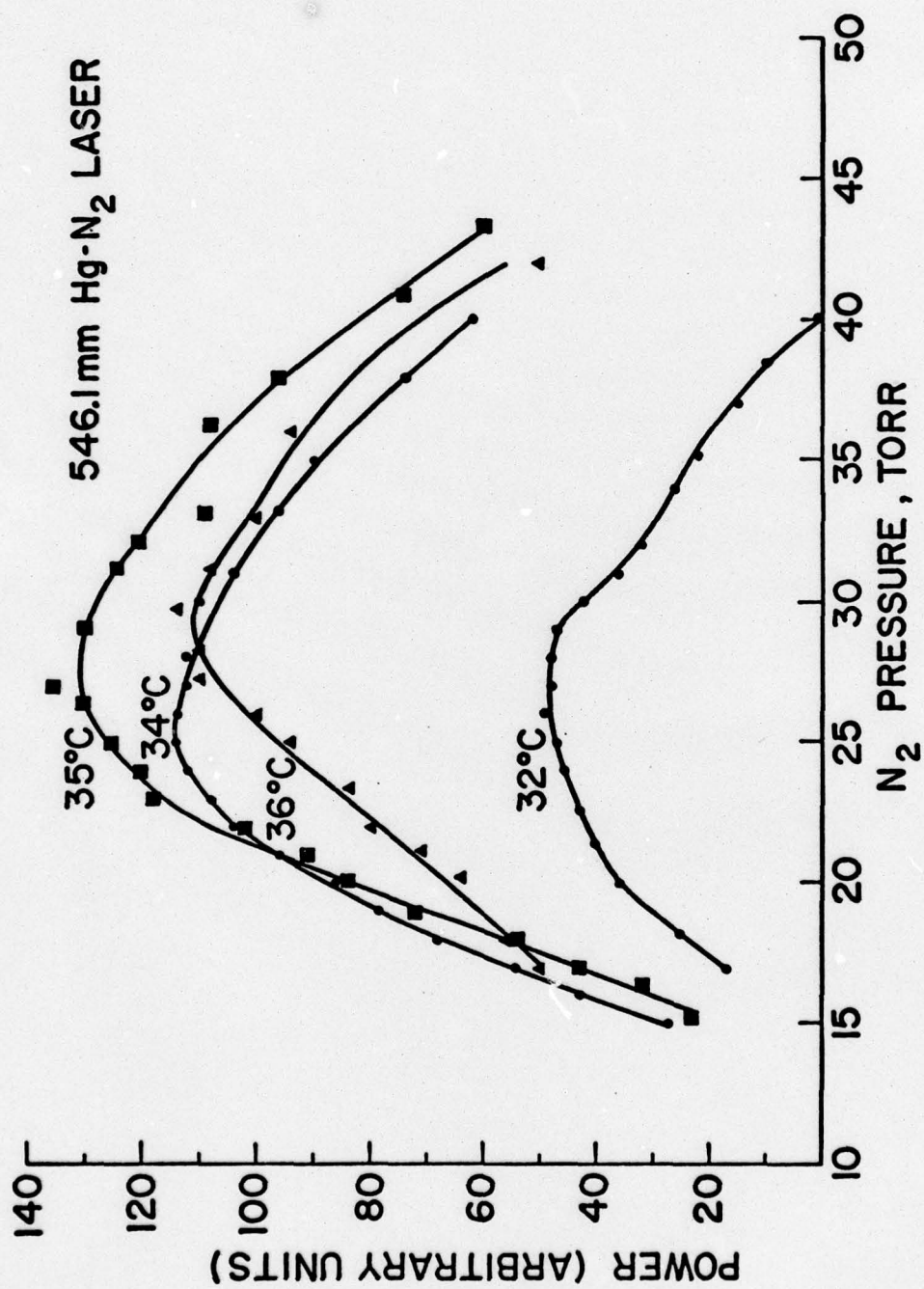


FIGURE 5

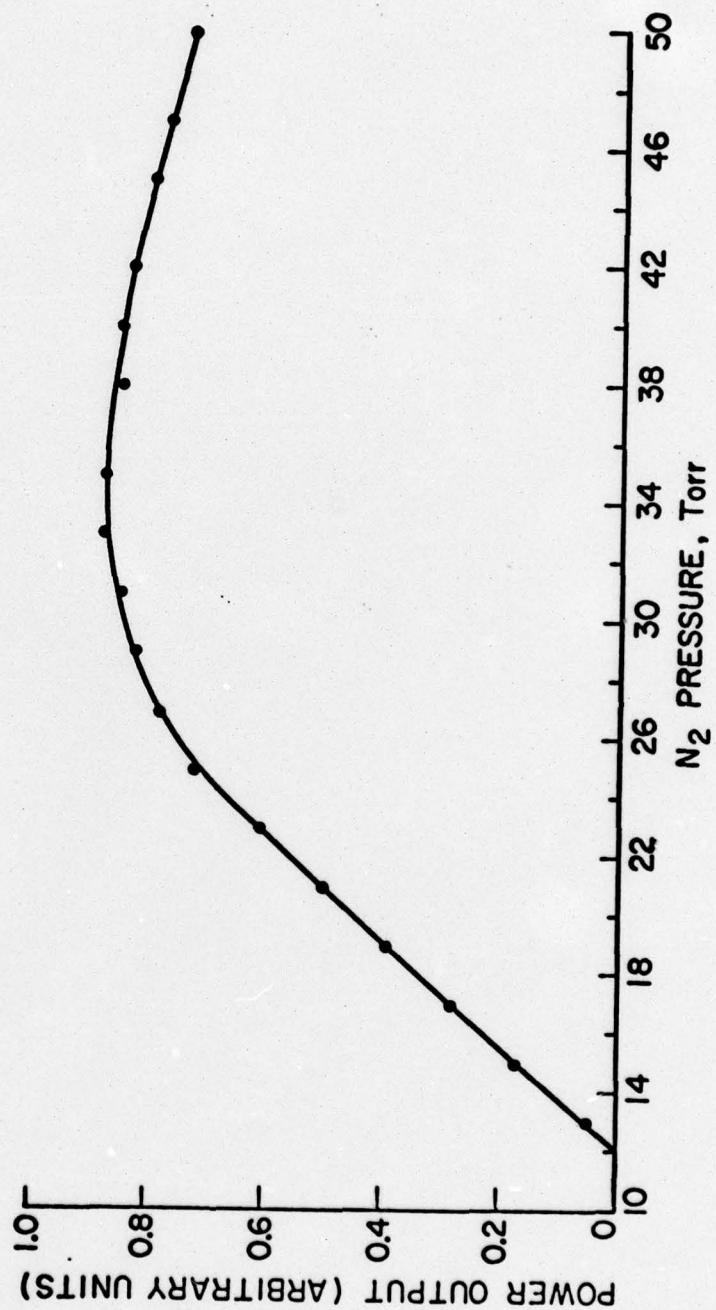


FIGURE 6

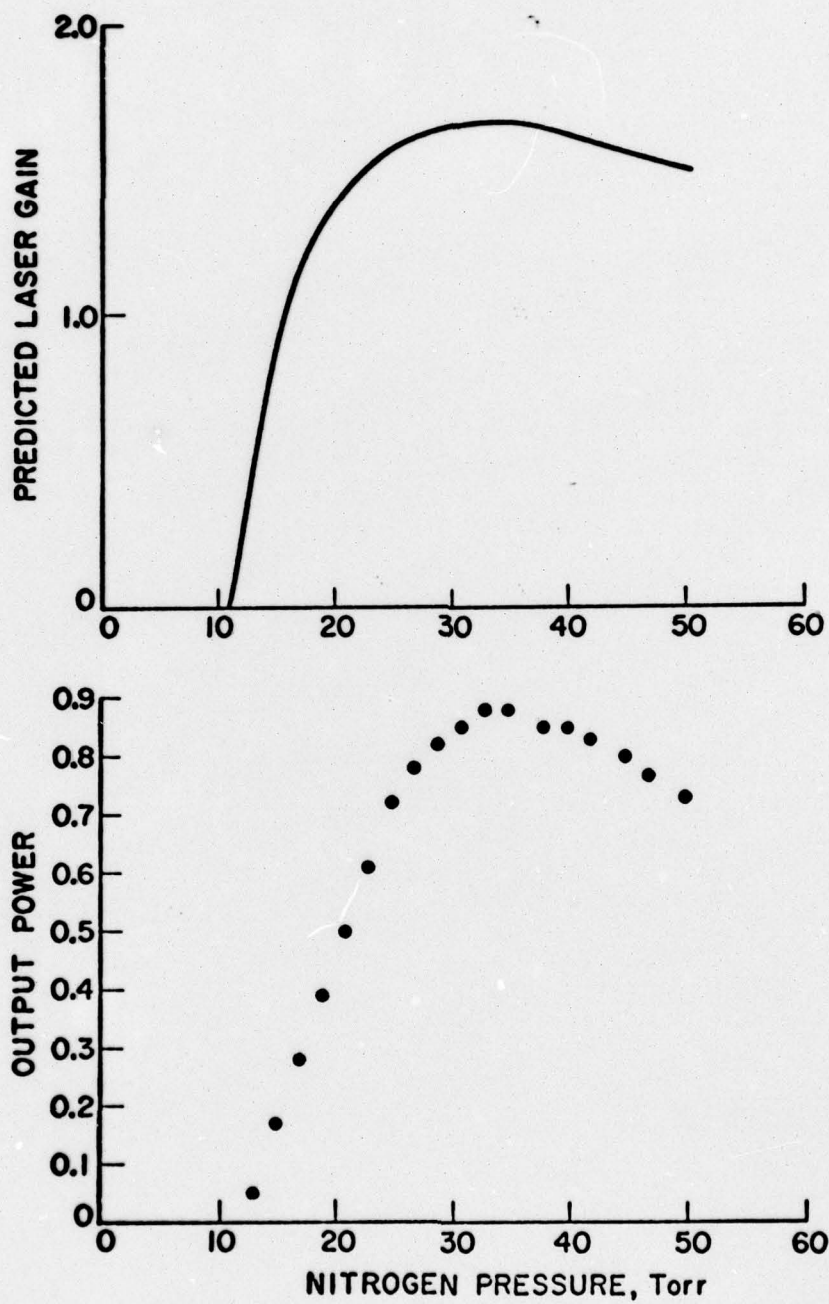


FIGURE 7

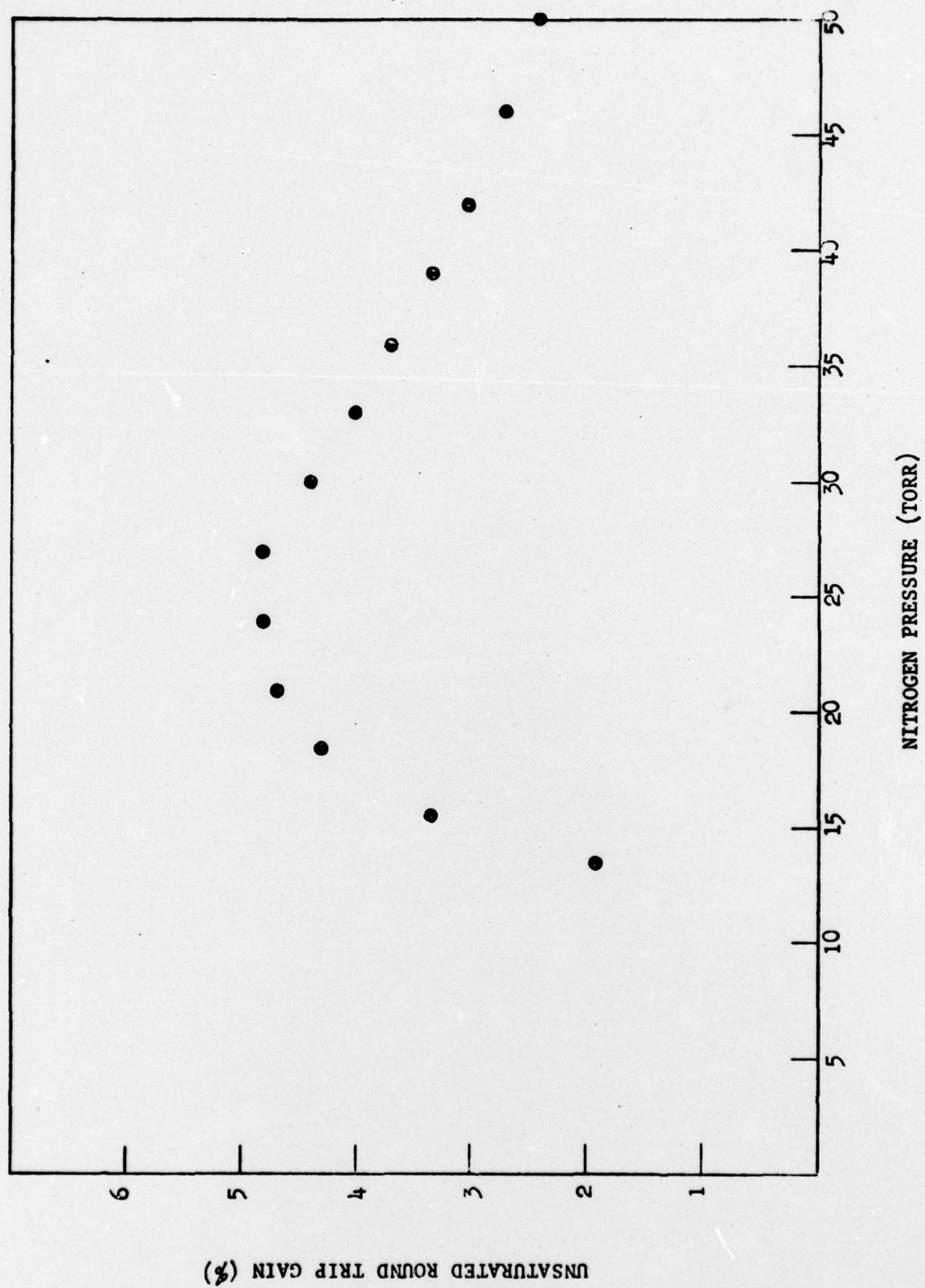


FIGURE 8

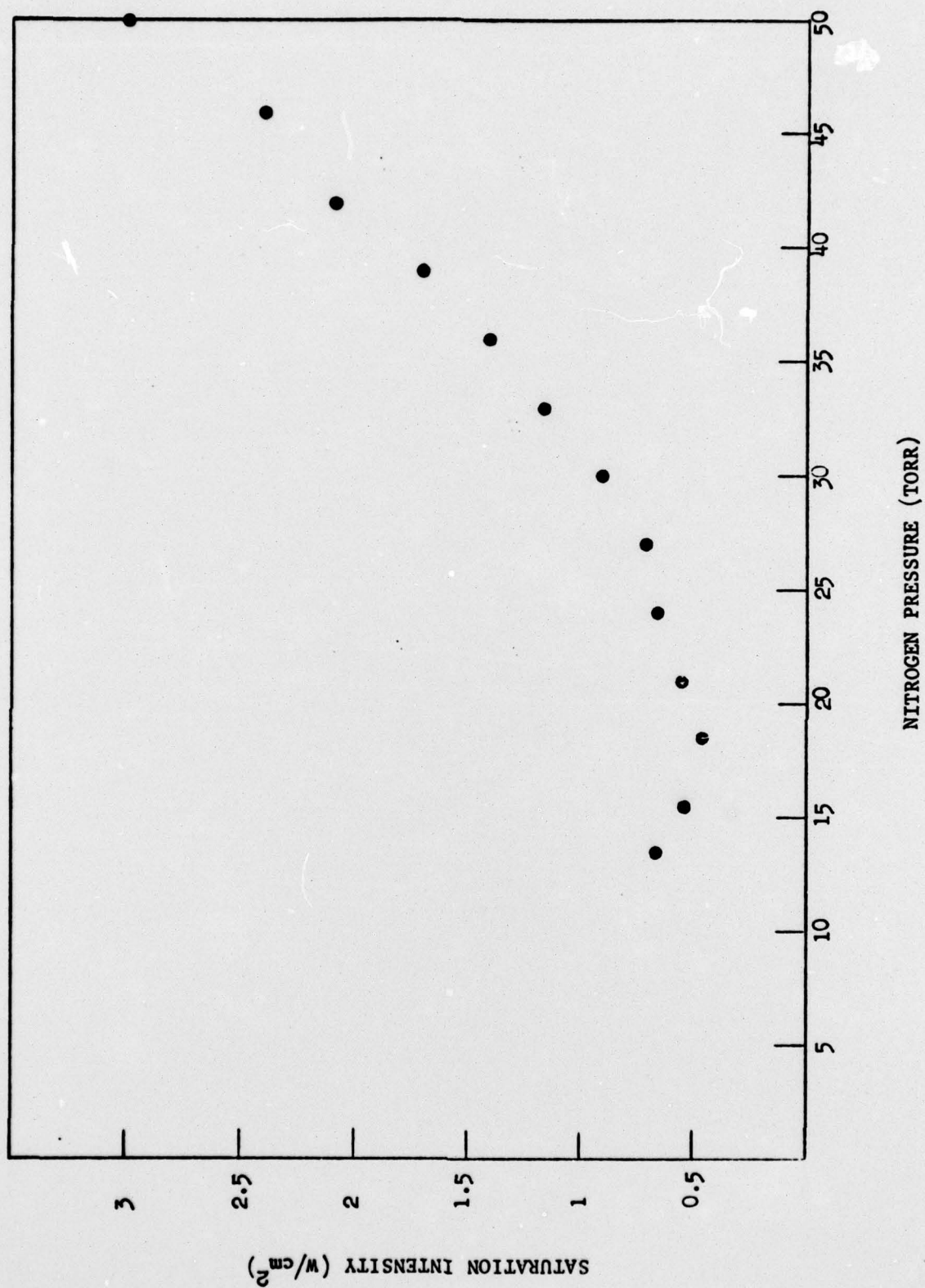
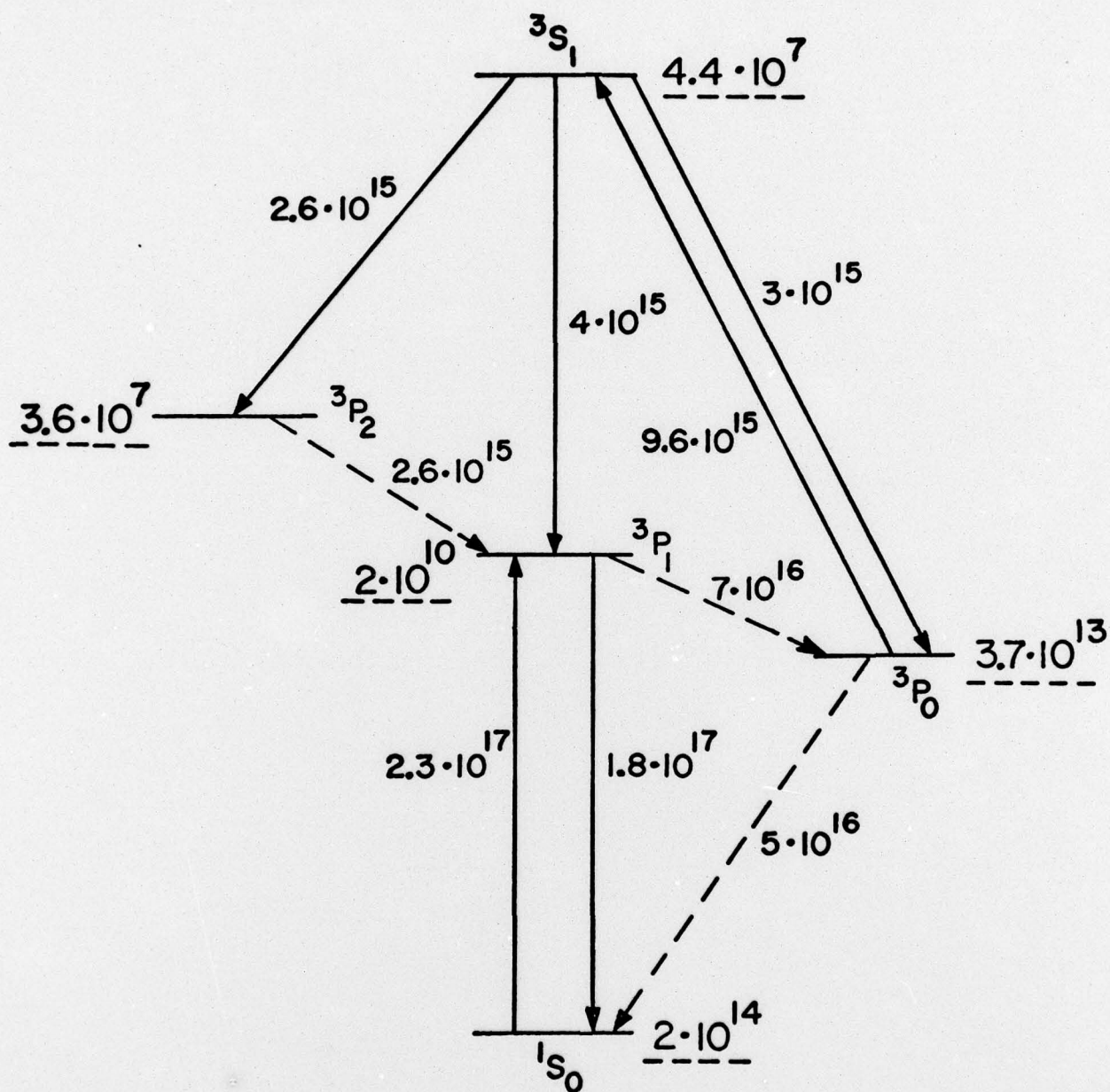


FIGURE 10

entirely consistent nor adequately complete. Certain important rates remain unmeasured, while contradictory results are given for others. We are able, however, by using best estimates for the important rates, to obtain very satisfactory agreement with such results as the measured pressure dependence of gain and power output in our lasers. Figure 11 illustrates some preliminary predictions for level populations and transition rates between levels in the Hg laser, while Figure 12 illustrates a predicted gain curve versus N_2 pressure. From this rate-equation analysis we have also been able to obtain a considerably improved theoretical understanding of some of the measured results reported in the previous literature. By carrying out additional measurements, particularly of absorption from the $\text{Hg } ^3\text{P}_0$ metastables in our laser tube, we expect to resolve most of the remaining uncertainties and obtain a complete and accurate description of the Hg laser system.

Our analysis of the laser itself has clearly shown that transverse spatial variations across the laser tube are very important and must be properly taken into account. Transverse diffusion and relaxation at the tube wall plays a dominant role in determining the steady-state population density for the $\text{Hg } ^3\text{P}_0$ metastables. Hence an expansion in Bessel-function diffusion normal modes is required, although the lowest-order such mode is by far the dominant term in the expansion under most conditions. Radiative imprisonment of the pumping wavelengths in the laser tube also plays a major role, and must be accounted for by a spatial analysis which extends the usual Holstein type of radiation-trapping analysis. We are confident that this analysis can be combined with the physical measurements described above to give a full picture of the laser.



PREDICTED LEVEL POPULATIONS AND FLOW RATES FOR
Hg-N₂ LASER AT 26 TORR N₂ PRESSURE

FIGURE 11

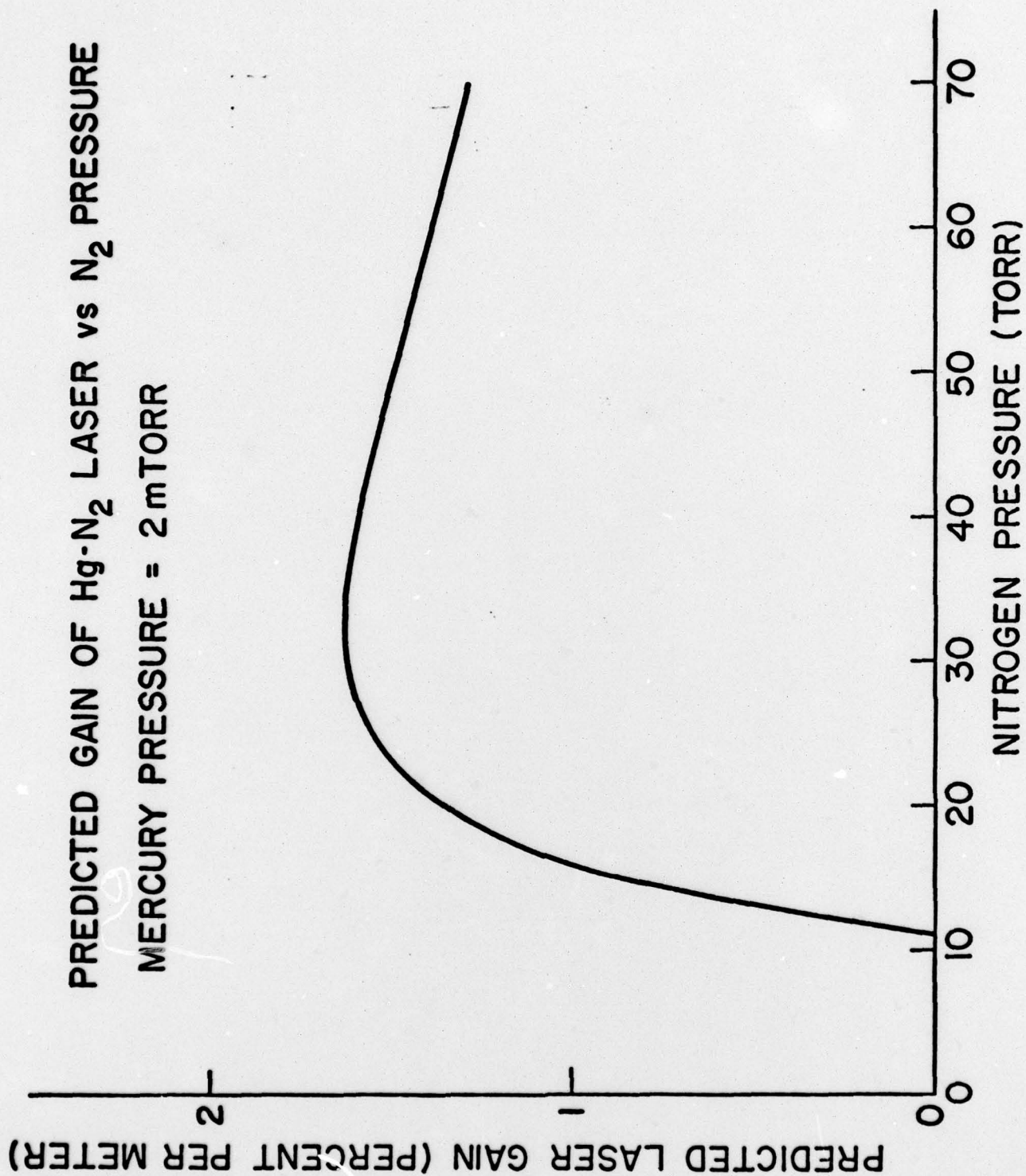


FIGURE 12

10. Alternative Gas Mixes

Nitrogen is almost unique among buffer gases in its ability to relax excited Hg 3P_2 and 3P_1 atoms into the desired metastable 3P_0 state without at the same time significantly quenching the excited Hg 3S_1 , 3P_1 , 3P_2 , and 3P_0 states back to the Hg 1S_0 ground state. Tabulations of quenching rates for other gases indicate that essentially every other molecule examined strongly quenches the Hg excited states and thus inhibits Hg laser action.

We have proposed and verified however that it is also possible to obtain weak Hg laser action by using as the buffer gas mixture a very low pressure of H_2O vapor, which serves to relax the Hg 3P_2 and 3P_1 levels to the metastable 3P_0 level, in combination with a larger pressure of either Ar or He whose primary function is to inhibit rapid diffusion and relaxation of the Hg 3P_0 metastables at the tube walls. Weak Hg laser action was obtained with buffer gas mixtures of Ar + H_2O , He + H_2O , and N_2 + Ar + H_2O . However, because this laser action is always inferior to pure N_2 + Hg mixtures, and because the required small H_2O vapor pressure is very difficult to control in practice, this discovery appears to be useful only for theoretical understanding and not as a practical technique.

It was also suggested to us (by Dr. C. K. Rhodes of SRI) that using N_2^{15} as the buffer gas might significantly improve the laser performance. The relative spacings of the Hg 3P_2 - 3P_1 - 3P_0 levels compared to the molecular N_2 vibrational levels are such that the desired Hg relaxation processes might be substantially faster with a slightly smaller molecular frequency. Use of the heavier N^{15} isotope in place of the usual N^{14} may accomplish this result. A supply of isotopically selected N_2^{15} has been obtained, and we expect to be able to try this experiment before the present research is concluded.

11. Mercury Laser Frequency Stabilization

One of the more promising applications for the mercury laser is as an absolute frequency-stabilized or wavelength-stabilized visible laser. Because there is no discharge in the laser tube itself both the spectral purity and the long-term stability of the mercury laser may be potentially very high. The wavelength of the mercury laser also falls in a spectral region where frequency stabilization by saturated absorption in molecular absorbers such as iodine may be very effective. The large number of mercury isotopes available increases the chance of finding a good overlap between the exact mercury laser frequency and a desirable molecular line for stabilization.

We have carried out a number of preliminary measurements that confirm the potential suitability of the mercury laser for wavelength stabilization and give some useful preliminary data. As a first test we verified that the beam from a free-running natural-mercury laser excited substantial fluorescence from cells containing Br_2 , I_2^{127} , and I_2^{129} . This indicates that there is at least some wavelength overlap between the Hg laser and absorption lines suitable for saturated-absorption spectroscopy in all three of these gases.

In a later experiment with an I_2^{127} cell placed inside the cavity of an isotopically selected Hg^{202} laser, a single strong saturated-absorption peak was observed in the laser's output power as the oscillation frequency of a single axial mode was tuned across the mercury transition linewidth using a piezo-scanned mirror on the laser. Figure 13 shows a tracing of the laser power output, including the saturated-absorption peak, on a single frequency scan. Because the laser cavity used in these experiments lacked adequate mechanical stability there was considerable frequency jitter on repeated

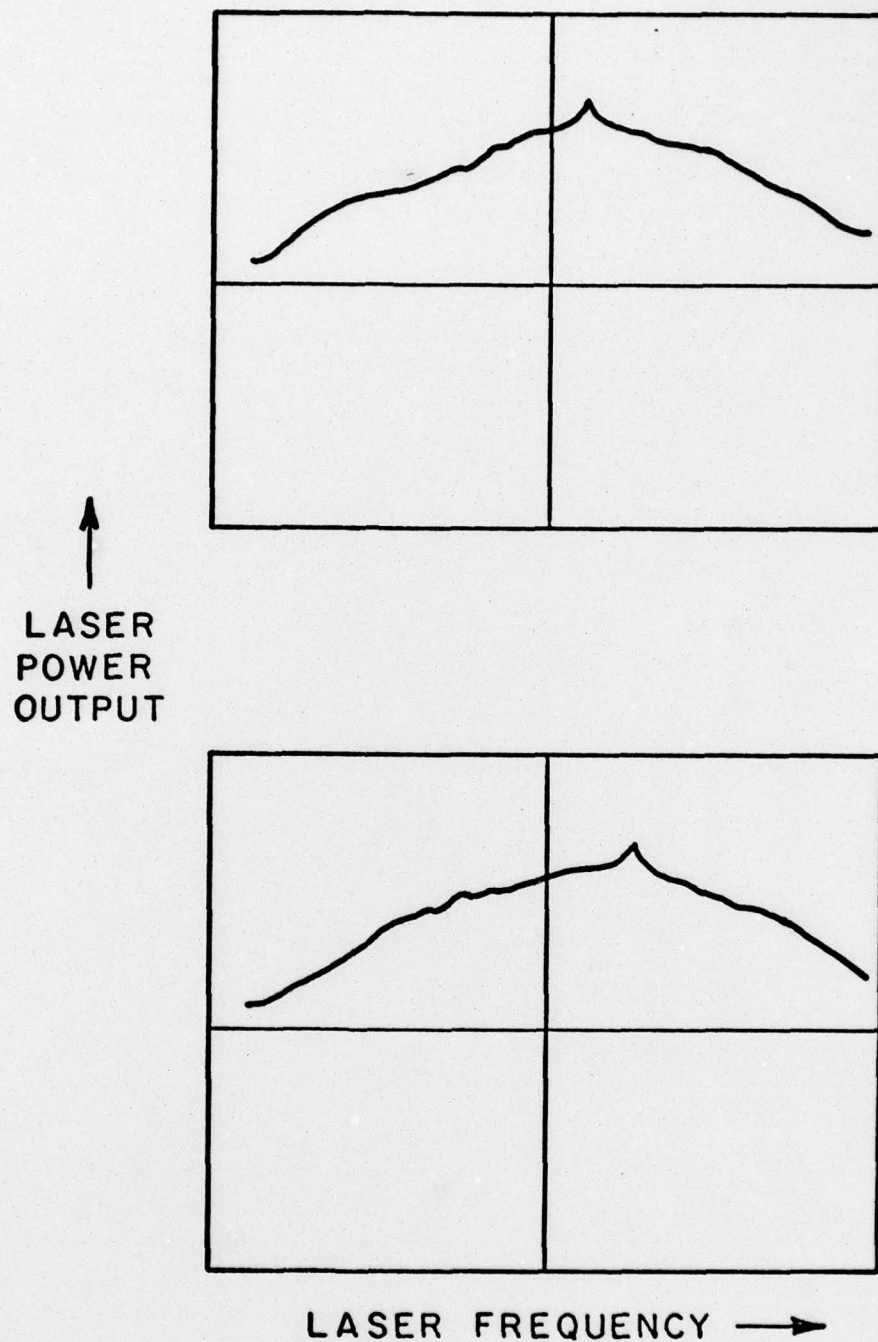


FIGURE 13

scans in these experiments, and it was not possible to stabilize the laser directly on the saturated-absorption peak. However, only a single such feature was observed with I_2^{127} over the entire available tuning range of the Hg^{202} laser. If the characteristics of this absorption feature turn out on further examination to be well suited to absolute stabilization, the presence of only a single such line will be a desirable attribute.

It was also observed that the relative position of this feature within the mercury laser's oscillation range changed as the N_2 pressure within the mercury laser was varied, indicating that the laser transition frequency was being pressure tuned across the frequency of the I_2^{127} absorption line. Figure 14 is a plot of the relative position of the I_2^{127} feature within the Hg^{202} laser's oscillation range of ~ 100 MHz as the N_2 pressure was varied. It should be noted that this is preliminary data: there is an uncertainty of as much as $\pm 20\%$ in the calibration of the frequency scale in this plot, and the absolute sign of the frequency deviation was not determined. However, it is clear that there is a pressure shift of the 546.1 nm transition in Hg^{202} with an absolute magnitude of $\Delta f_s/P \sim 9$ MHz/Torr. Operating the laser outside the pressure range shown in Fig. 14 moved the laser transition entirely off the I_2^{127} absorption feature, and no other such features were observed outside this range.

This measurement of the pressure shift Δf_s is significant in several ways. First, the pressure broadening coefficient of the laser transition, $\Delta f_a/P$, is almost certainly as large and possibly two to three times larger than the pressure shift coefficient $\Delta f_s/P$. Hence the pressure broadening of the laser transition in the 20 to 40 Torr operating range is probably 200 to 400 MHz, and perhaps larger, compared to a doppler linewidth of ~ 480 MHz.

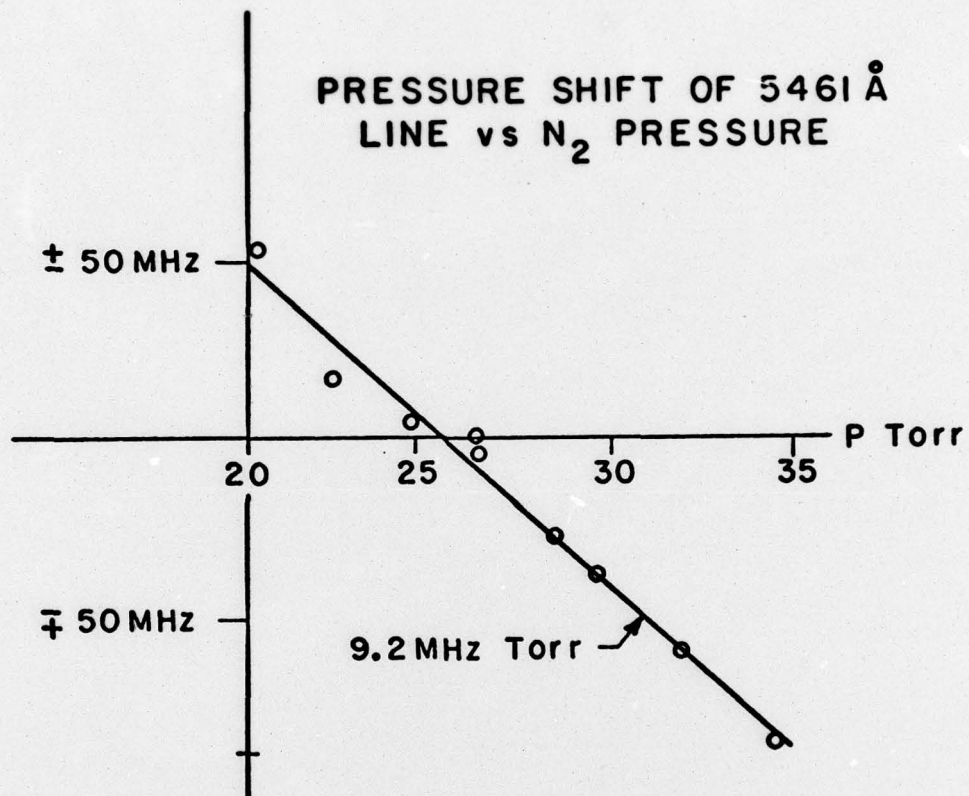


FIGURE 14

Hence, the laser transition lies somewhere between strongly inhomogeneous and homogeneous broadening. As a result, there is not expected to be any significant Lamb dip, in agreement with our observations. The laser can run in multiple axial modes, but the increased homogeneous broadening makes single-axial-mode operation somewhat easier at higher pressures. This is also in agreement with our observations.

Secondly, it is possible by controlling the N_2 pressure of the Hg laser within its central operating range to center the I_2^{127} absorption feature on the Hg^{202} transition. In a highly stabilized laser system this is desirable in order to minimize frequency pulling effects from the laser transition itself.

Thirdly, the possibility that the Hg 404.7 nm pumping line may have a pressure shift of similar magnitude has an impact on pump lamp design and evaluation for the mercury laser. If the 404.7 nm line in the low-pressure pump lamp is broadened but essentially unshifted, while the 404.7 nm absorption line in the laser is shifted with increasing pressure, this will need to be taken into account both in designing the pump lamp isotope mix and operating parameters, and in interpreting the pressure variation of laser performance in comparison with theory. Further work on this point will be carried out.

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20. ABSTRACT (Continue)

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